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PROVISIONAL APPLICATION FOR PATENT COVER SHEET

This is a r quest for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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INVENTOR(S)

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
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☐ Additional inventors are being named on the ______ separately numbered sheets attached hereto

TITLE OF THE INVENTION (500 characters max)

INTEGRATED STAGE FLOW THROUGH CAPACITOR

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ENCLOSED APPLICATION PARTS (check all that apply)

☑ Specification Number of Pages 10

☑ Drawing(s) Number of Sheets 7

☐ Application Data Sheet. See 37 CFR 1.76

☐ CD(s), Number

☑ Other (specify) Agreement/Assignment (4 pgs. Postcard; Check; Recordation Form Cover Sheet (1 pg. Dupl.

METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT

☑ Applicant claims small entity status. See 37 CFR 1.27.

☑ A check or money order is enclosed to cover the filing fees

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The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.

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☑ Yes, the name of the U.S. Government agency and the Government contract number are: Army Research Office/Contract No. DAAD1900C0448

Respectfully submitted,

SIGNATURE Leslie Meyer-Leon

Date 08/06/2003

TYPED or PRINTED NAME Leslie Meyer-Leon, Esq.

REGISTRATION NO. (if appropriate) 37,381

Docket Number: 0652-015US1

TELEPHONE 508-790-9299/Fax 508-790-1955

USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT

PROVISIONAL PATENT
0652-015US1

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of: Marc D. Andelman et al.

Docket No. : 0652-015US1

Title : **INTEGRATED STAGE FLOW THROUGH CAPACITOR**

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INTEGRATED STAGE FLOW THROUGH CAPACITOR

Background of the invention

In order to purify concentrated water, flow through capacitors need to be staged. The purpose of this invention is to improve the staging efficiency of flow through capacitors. Prior art flow through capacitors have individual, separate cartridge holders per stage. A dead volume exists between the capacitor layers and the inside walls of the cartridge holder or capacitor container. This dead volume mixes with the water which is purified in each stage, and decreases the resolution of the band of purified water, due to band broadening, as it moves from one stage to another. Therefore, a need exists for an improved flow through capacitor which has less mixing of purified or concentrated water with each other or with the feed water. One special use of the present invention is the low energy desalination of sea water. The present invention achieves that end by removing a little bit of salt at a time, at low energy, then, amplifying this signal by virtue of synchronous charging of multiple capacitor stages that share the same capacitive material, spacer, and optional charge barrier layers in order to sharpen bands of purified and concentrated water. This band sharpening method uses staging to refine or sharpen concentration bands, analogous to other staged methods such as chromatography, distillation, and absorption.

Summary of the Invention

The integrated stage design of the present invention eliminates the need, cost, and complication of connecting separate cartridge holders per stage by containing multiple stages within the same cartridge holder. This integrated stage design reduces band broadening due to mixing with of the band of purified water with purified water as it moves from stage to stage.

The integrated stage design of the present invention incrementally sharpens Purified and concentrated bands of feed solution as it flows from stage to stage. In individually staged capacitors of the prior art, a dead volume exists between each multilayer capacitor cell and it's cartridge holder. In the present invention, each stage shares common capacitor material layers. Only the current collectors, or, where electrodes are conductive enough not to need a current collector, the electrodes, are independently actuateable and electrically separateable from each other. Therefore, multiple stages may share the same cartridge holder, thereby eliminating the need for separate current collectors. Elimination of excess dead volume through integrated staging allows operation of the capacitor at lower voltage while, at the same time, obtaining a deep purification peak. The result of this is that the capacitor may be operated at lower, more energy efficient voltages. Operation of capacitors at lower, more energy efficient voltages is described in US patent 6,325,901B1. Due to the mixing effect, this method requires extra stages to operate, and may reach a lower voltage limit, as voltage is lowered when used with concentrated water, such as sea water. Therefore, a need exists for a method and system which allows the capacitor to operate more efficiently at lower, energy saving voltages.

A major difference in construction of the capacitor of the present invention from the prior art is that, in place of integral current collectors backing one or more electrode

layers, multiple current collectors are used, arranged sequentially along a fluid flow path, arrayed within the same cartridge holder.. These current collectors are made to be charged according to various sequences and times. The sequential charging of the multiple current collectors may either be done in series or in parallel, and in addition, within the context of either a series
or a parallel flow cell.

Brief Description of Figures

Figure 1 Integrated stage flow through capacitor and system
1 Controller or logic means
2 Capacitor power supply
3. Switch power supply
4 ground
5 switches, relays, or FETS.
6. cartridge holder or capacitor casing
7 Inlet
8 electrode, or electrode assembly.
9 outlet
10 flow path
11 individual current collectors
12 individual current collector lead
13 integrated circuit, programmable logic chip, digital input/output means, or printed circuit
14 cell assembly
15 sensor means

Figure 2
8 electrode, or electrode assemblies
10 flow path
11 individual current collectors
12 individual current collector lead
14 cell assembly
15 electrically insulating spacer or gasket between electrode or electrode assemblies 8
16 flow space or flow spacer

Figure 3
Integrated stage flow through capacitor
8 electrode, or electrode assembly
10 flow path
11 current collectors
12 individual current collector lead
14. Cell assembly
16 flow space or flow spacer
17 optional fluid or electrical gasket

Figure 4
Spiral wound integrated stage flow through capacitor

8. electrode or electrode assembly layers
current collectors
12 Individual current collector leads

Figure 5

Nested current collector flow through capacitor
8 electrode or electrode assembly layers
9 inlet or outlet
10 flow path
11 current collectors
12 individual current collector leads

Figure 6
Series or series array flow through capacitor
6 cartridge holder or capacitor casing
7 inlet or outlet
9 outlet or inlet
11 current collectors
12 individual current collector leads
18 series separators
19 high resistive series cell separator flow path

Figure 7

6 cartridge holder
8 electrode or electrode assembly
20 concentrated solution flow channel
21 purified solution flow channel
22 manifolded flow channels for concentrated solution
23 manifolded flow channels for purified solution.

Detailed description of figures

Figure 1 is a schematic of an integrated stage flow through capacitor and system of the present invention. Controller or logic means 1 and any additional digital input/output means 13 controls actuates switches, relays, or Field Effect Transistors (FETS) 5 in either a pre programmed or feed back controlled sequence using conductivity, pH, or other composition data, or flow, amperage, or voltage data supplied to logic means by sensor means 15. The multiple switching means 5 , which may also be an integrated circuit or chip with multiple outputs, can be triggered to ground 4 and thereby allow flow of current to current collector leads 12 into individual current collectors 11. Cell assembly 14 is made of electrode or electrode assembly layers 8 with flow spacer or spacers 16 (shown in figure 2), and individual current collectors 11.

Figure 2. Represents an detailed view of a flow through capacitor cell assembly 14 of the present invention showing optional electrical insulating spacers 15 insulating electrodes or electrode assemblies 8 one from the other. Electrode assemblies 8 are backed by individual current collectors 11 which are individually connected to individually actuated electrical leads 12.

Figure 3 Represents an integrated stage flow through capacitor cell of the present invention with individual current collectors 11, current collector leads 12, against electronically conducting electrode or electrode assembly 8. The electrode or electrode assembly 8 is spaced apart by flow space or flow spacer 16 with optional flow or series electrical isolation gaskets 14. Flow path 10 is shown cutting across the multiple current collectors 11 connected to individual electrical leads 12. The multiple current collectors shown in figure 3 may be thin enough to be wires, for example, less than two millimeters in diameter.

Figure 4. represents a spiral or concentric flow through capacitor of the present invention with electrodes or electrode assembly 8 , flow path 10 intercepting multiple current collectors 11 connected to individual electrical leads 12.

Figure 5. represents a nested integrated stage flow through capacitor single electrode assembly layer 8 with associated individual current collectors 11 and current collector leads 12 , flow path 10 and flow inlet our outlet .

Figure 6 represents a flow through capacitor of the present invention with a matrix series connection, where individual cells may be connected in series both horizontally and vertically. Only individual current collectors 11 and current collector leads 12 are show, with the addition of inter-cell series separators 18 . An optional high resistive flow path 19 is included for use when current collectors 11 are connected in series across the stack. Either a parallel or a series cell, not shown, would be placed between the end current

collectors 11. Where series cells are used, and where each of these cells is in turn connected in series, the resulting capacitor cell would form a series matrix, with resulting voltage upon charge of the matrix the multiple of the series voltage of the individual cells time the number of series cells further connected together in series. Cartridge holder or capacitor casing 6 contains the matrix connected flow through capacitor formed by these components plus additional electrodes or electrode assembly layers 8 and flow spacers 16 shown in figure 2

Figure 7 represents a flow through capacitor of the present invention with the flow parallel to the current collector 11, which may be used to provide continuous concentration streams 20 and purifications streams 21. Concentration stream manifold 22 and purification stream manifold 23 combines together the concentration streams 20 and purification streams 21 produced by multiple electrode or electrode assembly layers 8.

Detailed Description of the preferred embodiments of the invention.

The integrated stage flow through capacitor of the present invention utilizes multiple current collectors 11 individually connected to current collector leads 12 with common flow path layers 14, and electrode or electrode assembly layers 8 that may be optionally separated by inter electrode insulators 19. The current collectors of the present invention are multiple in the sense that multiple current collectors are transected by a common flow path 10, share common electrode or electrode assemblies 8, or are placed within the same cartridge holder or capacitor casing 6. A stage is represented by any pair of current collectors typically perpendicular to, but optionally parallel to, the flow path 10. Flow path 10 therefore typically intersects oppositely charge current collector pairs. As the fluid in flow path 10 flows across these current collector pairs, the current collectors, through individual current collector leads 12, are activated in a sequence. This sequence may either be programmed or may be trigged by a sensor 17. Each time a pair of current collectors is activated with a voltage, additional materials are either removed from or added to a band of water which flows from cell to cell, where cell may be defined as any oppositely charged pair of current collectors charged together in time as part of a charge sequence among many pairs of current collectors. The multiple current collectors 11 of the present invention within each cartridge holder may bracket a stack of electrode assemblies 8 that are electrically insulated from each other in true series fashion, such as in WO0113389A1, or the current collectors may be bundled together in parallel, to form anode and cathodes, with geometries as in any prior art flow through capacitors including but not limited to the flow through capacitors below

PATENT #	TITLE
5,192,432	Flow Through Capacitor
5,196,115	Controlled Charge Chromatography System
5,200,068	Controlled Charge Chromatography System
5,360,540	Chromatography System
5,415,768	Flow-Through Capacitor
5,538,611	Planar,Flow-Through,Electric,Double Layer CapacitorAnd A Method Of Treating Liquids With The Capacitor
5,547,581	Method Of separating Ionic Fluids With A Flow Through Capacitor
5,620,597	Non-Fouling Flow-Through Capacitor
5,748,437	Fluid Separation System With Flow-Through Capacitor
5,779,891	Non-Fouling Flow-Through Capacitor System
6,127,474	Strengthened Conductive Polymer Stabilized Electrode Composition And Method of Preparing
6,325,907	Energy And Weight Efficient Flow-Through Capacitor, System And Method
WO 93/13844	Chromatography System With flow-Through Capacitor And Method
WO 94/26669	A Planar,Flow-Through,Electric,Double Layer CapacitorAnd A Method Of Treating Liquids With The Capacitor
WO 95/21674	Flow-Through Capacitor And Chromatogrphic System And Method
WO 97/13568	Non-fouling,Flow-Through Capacitor,system And Method Of separation
WO 00/14304	Flow-Through Capacitor And Method Of Treating Liquids With It
6,413,409	Flow-Through Capacitor And Method Of Treating Liquids With It
WO 01/09907 A1	Flow-Through Capacitor And Method
CN 00812144.3 (Appl)	Flow-Through Capacitor And Method
WO 01/13389 A1	Flow-through Capacitor system And Method
WO 01/66217 A1	Low Pore Volume electrodes With Flow-Through Capacitor And energy Storage Use And Method
WO 01/95410 A1	Fluid and electrical connected flow-Through Electrochemical Cells,System And Method
WO 02/29836 A1	Fringe-Field Capacitor Electrode For Electrochemical Device

In addition to the stacked layer geometries shown here, individual electrodes shown in the electrode array WO 03/009920 A1 may be sequentially charged according to the present invention in order to form flow through capacitor stages according to the present invention.

The electrode assembly 8 may include any combination flow through capacitor electrodes and optional current collector, ion perm selective membranes, or charge barrier materials known to the prior art, as described in. International Application No. US01/12641, "Charge Barrier Flow-Through Capacitor". For example, the electrode may be comprised of high capacitance particles, including carbon or ceramics, held together with any binder material including a hydrogel or functional charge barrier material, fibrillated polypropylene or PTFE, latex, or cross linked or sintered polymer or hydrocarbon materials. For example, if a charge barrier material is used as a binder, it may be a fluorine containing ionomer, an elastomeric or thermoformable hydrogel containing either or both of strong acid or strong base functionality.

Description of the embodiments.

The essence of this invention is the ability to vary the voltage either perpendicular or parallel to the flow path within a single cartridge holder. This is accomplished by incorporation of incorporating multiple current collectors within a single cell. These current collectors may have individual electrode, charge barrier, and flow spacer layers, or may share one or more of these layers in order to simply manufacture. A fluid, solution, or solute to be purified of concentrated is introduced into this cell. As this fluid flows through the cell, the multiple current collectors are independent ally synchronously actuated with a purification or a concentration peak that forms as the flow moves through the cell. This is similar to chromatography. For example, a small amount of total dissolved solids may be purified from water each time the flow stream passes a particular current collector, or pair, or set of current collectors. This produces a slightly purified aliquot of water. As this aliquot moves along it's flow path, additional current collectors are actuated. Where this flow path cuts across multiple current collectors, the flow is separated into purification and concentration peaks that may be separated by a valve. Where the flow is parallel to the multiple current collectors, the fluid will be separated into side by side purification and concentration streams.

Where electrode layers are shared between multiple current collectors, it is important that the current collectors are electrically isolated. To do this, the distance between current collectors has to be wide enough so that the current collectors have a fair degree of electrical isolation between them. Generally, wider than one sixty-fourth of an inch, and optimally, under three inches, such as between one eight and one quarter inch, will suffice to isolate the cells. Cells are sufficiently isolated when the width between current collectors backing an adjacent electrode material of a given conductivity is such that the overall conductivity of the combined current collector – electrode combination is 1 ohm cm or greater. Resistance between individual current collectors are preferably 0.1 ohm or more.

US patent 6,325,901B1 teaches that operation of a flow through capacitor at lower voltages has an energy advantage. However, this application reaches a practical limit due to the fact that less total dissolved solids and other contaminants are removed from water per charge cycle as voltages are lowered. The flow rate utilization per gram of carbon of this prior art capacitor needs to be lowered in order to provide a given percent purification at a lowered voltage and energy usage level. This slowing of the flow rate is necessary in the prior at because sufficient levels of purification must be maintained in order to overcome the mixing effects of the cartridge holder dead volume. The capacitor of the present invention minimizes this cartridge holder dead volume by eliminating inter-stage cartridge holders. For example, with integrated staging, the dead volume due to the flow spacer may be larger than the dead volume due to the volume that exists between the capacitor cell and the inside of the cartridge holder. The capacitor cell is defined as the electrode, flow spacer, and any current collector and charge barrier layers. Therefore, focused bands of purified water may be obtained at low voltages and therefore low energy usage levels. An advantage of the present invention is that energy may be saved by charging the integrated stage cell many times at lower voltages. Percent utilization of the cell may be increased by virtue of operating the cell so that more than one concentration band is present at any given time within a given material layer or cartridge holder. To obtain purified water at a desired purification level, a valve coupled with a conductivity sensor selects out water desired purification and concentration levels. Percent recovery may also be preselected by presetting conductivity control based upon a mass balance between purified, feed, and/or waste concentrations.

Continuous purification

An additional advantage of the present invention is that it allows operation without a valve. In this case, flow may be parallel to the current collectors, and fluid concentration may be modulated within the plane of the flow. Figure 7 depicts how water may be purified 21 between one set of current collectors and concentrated 20 between adjacent sets of current collectors, with purification streams 21 and concentration streams 20 are coplanar in the direction of flow. These coplanar streams are manifolded together to form a continuously purified stream 23 and concentrated stream 22. Optionally, ions may be manipulated by operation of the current collectors in order to form a central flow path of one concentration and side flow paths of another. In either case, adjacent concentration bands are formed which may be directed to separate

collections paths or separately collected from separate outlets in the cell casing provided for this purpose. A valve is not except when it is desirable to switch concentration and purification streams by polarity reversal or otherwise electronically manipulating the charge cycle sequence.

Individual current collectors may be pre manufactured onto electrode layers by lamination or by continuously coating a banded pattern of current collector material onto an electrode material. The current collector material may be any conductive material less than 1 ohm cm in resistance, including a graphite material, or, may be a metal foil, in particular, metal foil protected by a conductive vinyl or any form of carbon-polymer composite layer using carbon black, nanotubes, graphite powders, etc. mixed in with a thermoplastic, olefin, fluorocarbon, or any other polymer, used with a metal backing to form a current collector where the metal is protected from water by the thin polymer layer, with the polymer layer typically under .03 cm thick. Conversely, a capacitance containing electrode material may be discontinuously formed in a banded pattern upon a suitably conductive current collector layer. Any carbon containing material may be used. Microparticulate titanium oxide powder is also advantageous.
Electrode materials may include carbon black, activated carbons, sintered carbons, aerogels, glassy carbons, carbon fibers, nanotubes, graphite, and other forms of carbon in particular those with surface areas over 400 BET, nanotube gels, conductive polymers, ceramics, anatase titanium oxide, or any other material that has a capacitance above 1 Farad per square meter when applied in a layer 0.1 cm or less thick. Any binder material may be used which holds the electrode capacitive particles together, including hydrogels, latex, thermoplastics, and fluropolymers.. Charge barrier materials may be any ion exchange polymer, hydrogel, cross linked polymer, or membrane, preferably with a charge density of over 0.1 milliequivalents per gram. The charge barrier material may also function as the binder or adhesive which is used cause capacitance containing electrode particles together to form an electrode sheet, or to adhere them to a current collector..

To manufacture the present invention, current collector, electrode, and any charge barrier layers may be laminated or layered together in a stacked configuration, then cut to length and inserted into a cartridge holder. Only as many layers as may be effectively cut at one time are layered and pre cut together. The blocks of materials so formed may in turn be layered on top of one another and inserted into a cartridge holder designed to fit them. Current collector tabs may overlap out either end, and be bundled together in parallel or connected in series. If graphite foil is used as a current collector, the end tabs may be infiltrated with an oily material or a polymer so that they do not wick water. A metal compression contact may be formed with a nut , bolt, and-or washer arrangement to the bundled graphite tabs. This contact may be protected from water by a compression nut containing a gasket which screws down over the top in order to cover underlying metal contacts. Inert metals such as titanium, tantalum, including palladium or platimum coated or infused valve metals, may be used to form metal to graphite contacts. It is preferable that contact resistance be less than 1 ohms. Metal contact to graphite compression over 10 psi, and metal to graphite contract areas over 1 cm^2 may be used to achieve this.

Claims

1. A flow through capacitor for the purification of solutions where one or more the flow spacer, charge barrier, or electrode layers are shared among multiple current collectors.

2. The capacitor of claim one which is operated such that multiple concentration bands exist simultaneously within a given material layer

3. The flow through capacitor of claim 1 whereby the flow is across multiple current collectors, and desired purification and concentration streams are selected out by means of a conductivity controlled valve

4. the flow through capacitor of claim 1 whereby flow is parallel to the multiple current collectors, with continuous purification and concentration streams directed to separate collection paths.

5. the flow through capacitor of claim 4 whereby fluid is manipulated to form adjacent purification and concentration streams that may be separately collected without need for a valve.

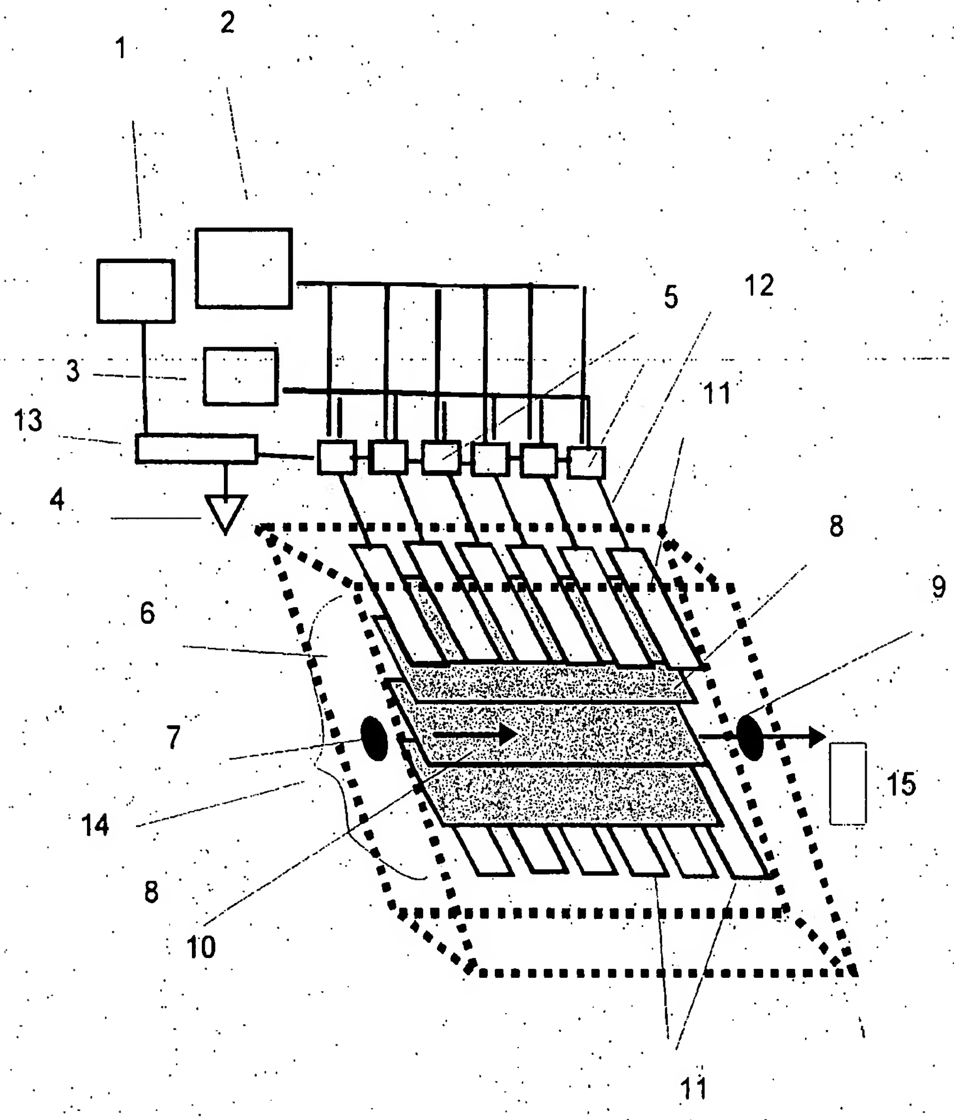

Figure 1

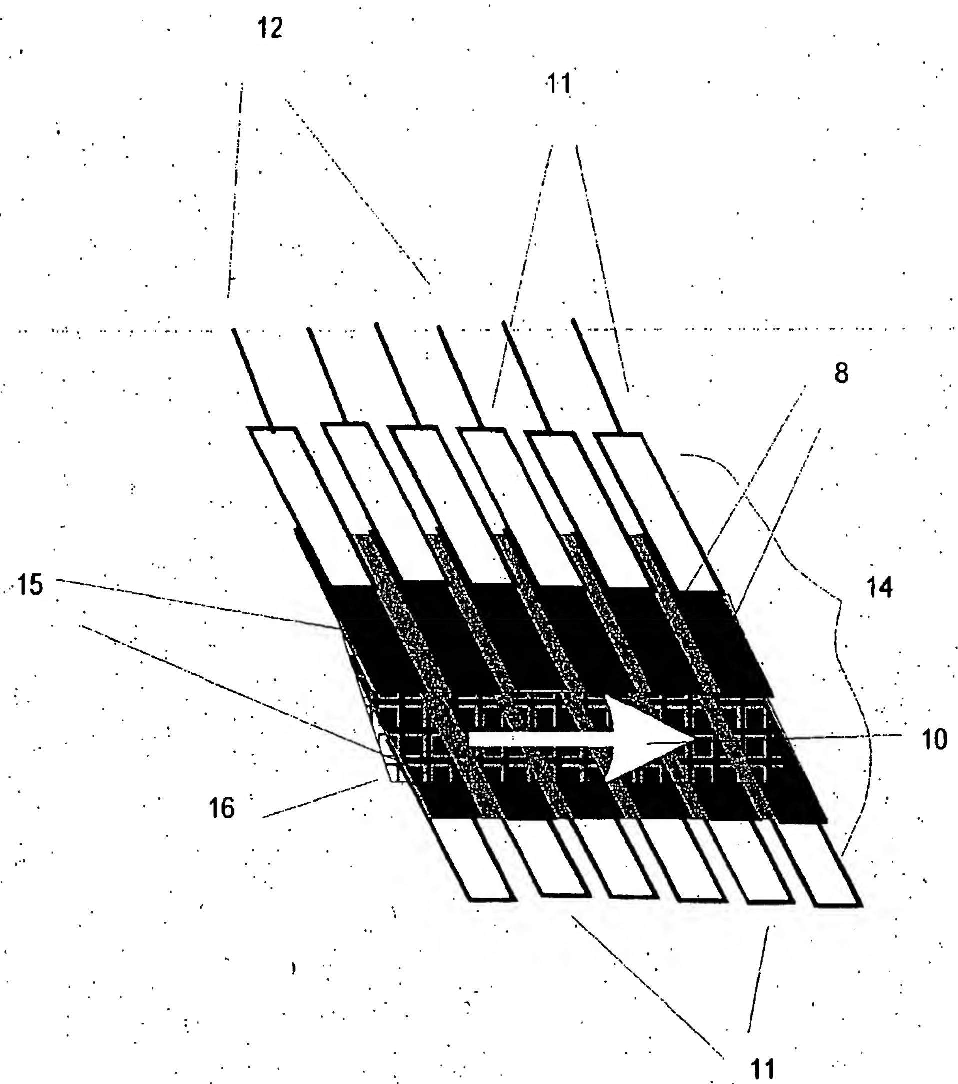

Figure 2

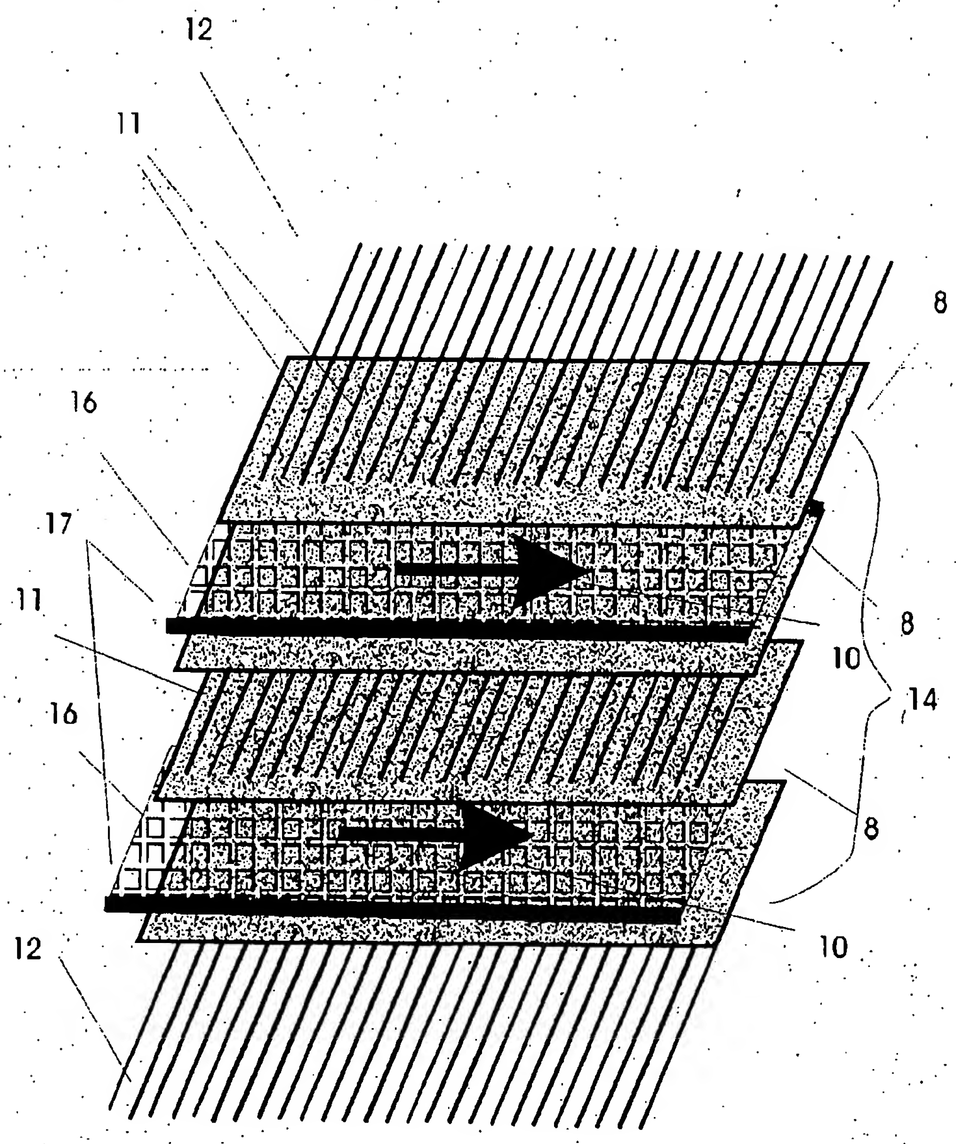

Figure 3

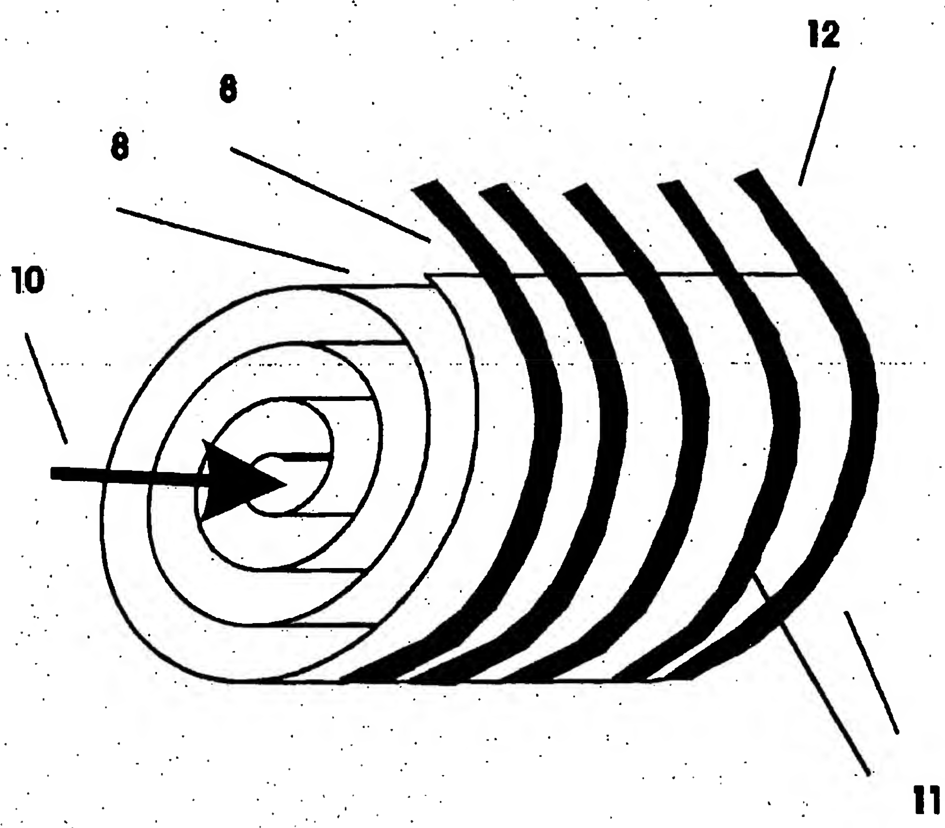

Figure 4

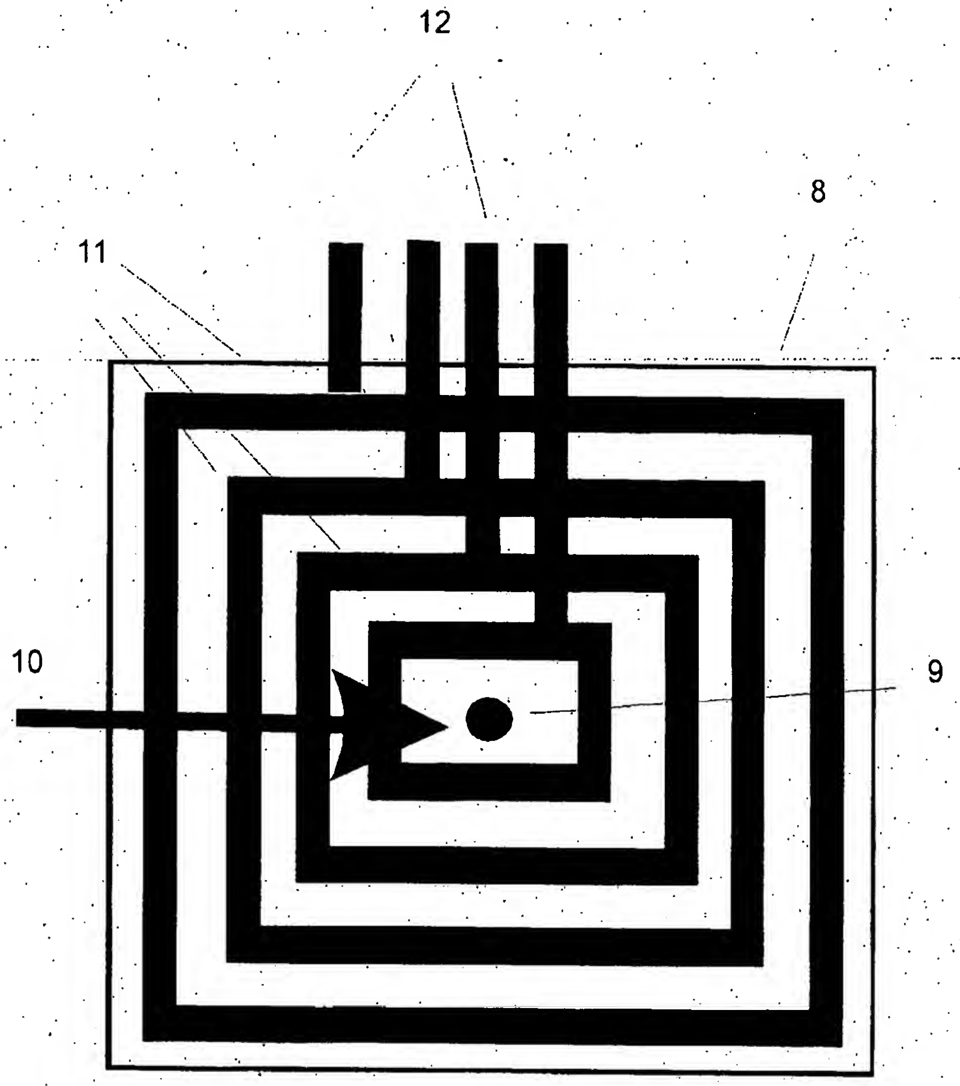

Figure 5

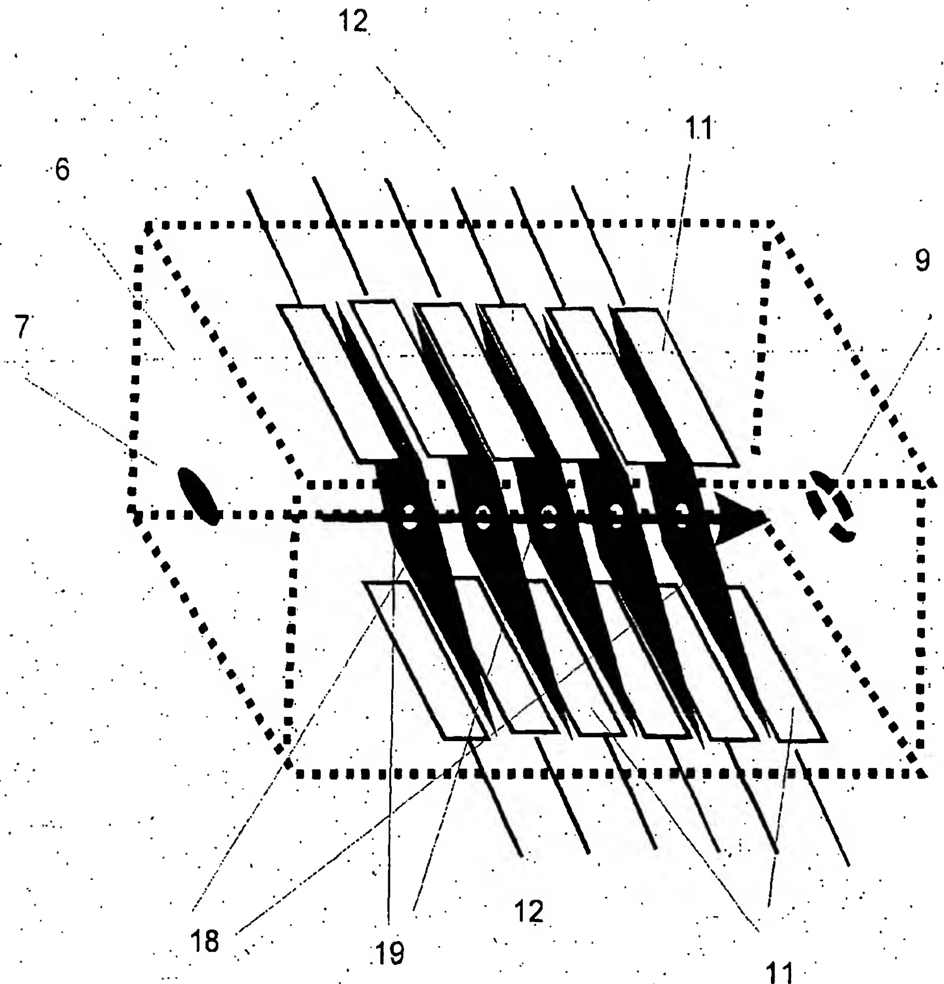

Figure 6

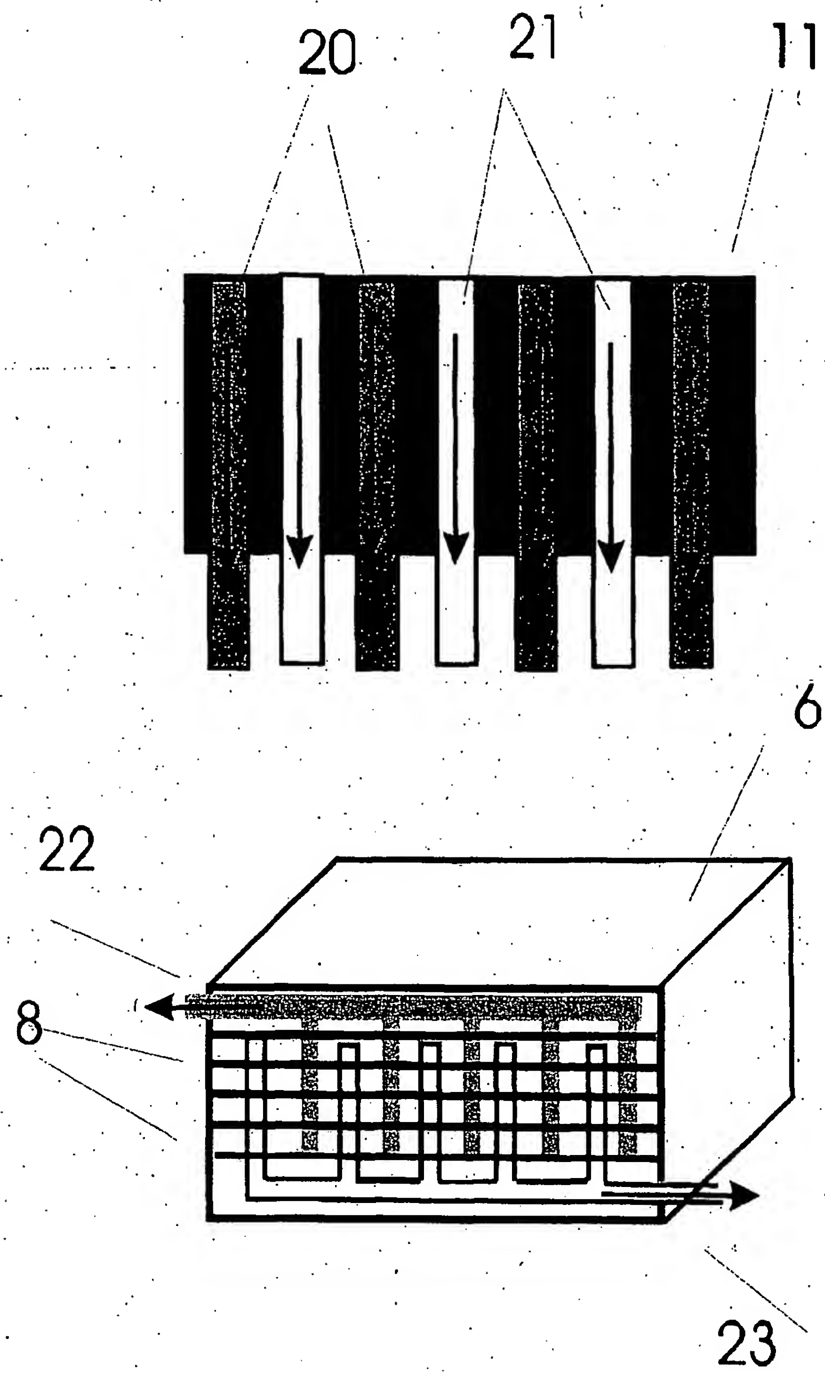

Figure 7